



AP  
1700

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES**

In re application of:

Nilo FAGIOLINI et al.

Serial No. 09/423,746

Filed: November 15, 1999

For: REACTIVE POWDER COMPOSITION  
AND METHOD FOR PURIFYING GAS

Art Unit: 1754

Examiner: T. Vanoy

Atty. Docket No. 32232-15219

Customer No.



26694

PATENT TRADEMARK OFFICE

GROUP 1700  
APR 17 2003  
RECEIVED

#32/UB

4/28/03

**SUPPLEMENTAL BRIEF**

Assistant Commissioner for Patents  
Washington, D.C. 22031

Sir:

The SUPPLEMENTAL BRIEF is filed in response to Paper No. 29. In Paper 29, the Examiner required augmentation of the Appeal Brief filed Dec. 2, 2002

- 1) to insert a reference to the drawings
- 2) to insert positive indication of that Claim 3 had been cancelled "in the 116 Amendment date-stamped Sept. 18 2002, and the "Status of the Claims" and "issues" portion of the appeal brief"
- 3) to correct Claims 6 and 13; and
- 4) to refile an IDS.

The Supplemental Brief is filed in triplicate and concurrently with an IDS [as requested by the Examiner and discussed below] and an Amendment.

Moreover, in Paper No. 29, the Examiner has asserted against Claim 13 a ground of rejection under 35 U.S.C. 103 which had been lodged against claims 1, 2, 4-12 [over German Patent No. DE 4100645 (Regler) taken with WO 95/19835]; please see page 6 of Paper No. 17.

The additional grounds of rejection of Claim 13 and items 1)-3) is addressed in this paper. The references filed on October 1 and twice on December 2 2002 are presented under separate cover letter with the heading INFORMATION DISCLOSURE STATEMENT.

In Paper No. 29, the Examiner indicated a 3-month shortened Statutory period from the 1/16/03 mail date of Paper No. 29 for filing a SUPPLEMENTAL BRIEF.

The SUPPLEMENTAL BRIEF is presented within the spirit of the MPEP Section 1208. Brief fees were previously submitted, according to the file in the undersigned office.

References relied upon by the applicants during prosecution are attached hereto, including copies of

**Kirk Othmer Encyclopedia of Chemical Technology**, Fourth Edition, volume 22, Wiley Interscience Publication, p.276. [1997], including copyright page.

a page of Terence Allen's book entitled "Particle size Measurement", Second Edition, published by Chapman and Hall Ltd.(1975)[Enclosure 1] and

a copy of a page from Ullmann's, Encyclopedia of Industrial Chemistry, Fifth Edition, Vol. A7, p. 160-161, 1986, editor Wolfgang GERHARTZ, publisher: VCH., (Enclosure 2)].

These references were attached to the two responses to the FINAL REJECTION.

#### **REAL PARTY IN INTEREST**

The instant application is assigned to SOLVAY S.A.

#### **RELATED APPEALS AND INTERFERENCES**

The undersigned is not aware of any appeal or interference which would affect, or be affected by, the outcome of the appeal in this case.

#### **STATUS OF CLAIMS**

The claims 1-2, 4-10, 12 and 13 stand finally rejected under 35 U.S.C. §§103 and/or 112 and for obviousness type double patenting. (Claim 3 was cancelled after the Final Rejection.)

#### **STATUS OF AMENDMENTS**

The initial Rule 116 September 3, 2002 AMENDMENT was denied entry [PTO Paper no. 21]. Resubmission of the amendment with conformance of the 'clean copy' to the 'marked up version' of amendment of Claim 1 therein resulted in entry of the September 18, 2002 Rule 116 AMENDMENT, as set forth in PTO paper no 24. A paper entitled "**PRESENTATION OF EXCERPT FROM KIRK-OTHMER**" was filed by applicants on October 1, 2002, was refiled with the original Brief, and now per Paper No. 29, is filed with the title INFORMATION DISCLOSURE STATEMENT. Concurrently with this SUPPLEMENTAL BEIF, an AMENDMENT under RULE 116 is presented. (The Amendment proposed is of Claim 13), which proposal is reflected at page 17.

#### **SUMMARY OF THE INVENTION**

The invention concerns a solid pulverulent reactive composition based on sodium bicarbonate, for gas purification and scrubbing gases which may contain sulfur oxides and/or hydrogen halides [please see specification at page 1 line 10 et seq.].

The composition comprises

sodium bicarbonate and

a caking inhibitor for sodium bicarbonate;

said inhibitor is selected from the group consisting of lignite coke, a magnesium compound and admixtures thereof, wherein said magnesium compound is selected from the group consisting of magnesium oxide, magnesium hydroxide, mixtures of magnesium oxide and magnesium hydroxide and magnesium hydroxycarbonate;

The composition of the claims is said to be devoid of silica. The invention addresses the problem of agglutination of such compositions, the tendency of the sodium bicarbonate to cake). The inventors have discovered that silica content in such compositions negatively affects the gas cleaning process, in particular when the removal of dust is carried out by means of a filter cloth (Please see the specification at p.5, 1. 24-34). The inventors believe that the silica-free compositions adhere better to the filter cloth than the silica-comprising compositions. The reference to filter clothes relates to means used to remove dust, **i.e. solid waste**, from the gases. Please see page 5 line 15 et seq. of the specification relating to dust removal. Accordingly, Claims 1, 12 and 13 recite "devoid of silica".

Moreover, according to a specially recommended embodiment of the invention composition, which adheres well to such filter cloth, the composition is characterized by a fine mean particle size of less than 50 $\mu$ m and a narrow particle size distribution (slope of less than 5) (see description p.3, 1.19 – p.4, 1.4).

Such particle sizes can be obtained by milling commercial sodium bicarbonate (see example 1). Amended claim 1 incorporates the characteristics of particle size: "*said composition exhibiting a mean particle size of less than 50 $\mu$  and a particle size slope of less than 5.*" The particular particle sizes of the composition according to new claim 1, in conjunction with the new caking inhibitor and the absence of silica, have proven to be particularly effective, as demonstrated by the examples of the patent application.

Such particle sizes are not those of standard sodium bicarbonate but require additional treatment, for instance milling and sieving, as described in example 1 of the specification at page 8.

Figure 1 and Figure 2 are noted, in view the Examiner's express request at page 3 paragraph 1) of Paper No. 29. Figures 1 and 2, respectively, relate [1] to the array of storage units of compositions of the invention and subsequent testing and [2] to the equipment for, measuring mobility [the ability to flow freely, as described at page 7 line 17 et seq] of the reactive composition of the claims, after storage. Figure 1 illustrates the array of stacking of bag units of the composition. Figure 2 relates to the device comprising a sieve 9, exhibiting a mesh size of 710  $\mu\text{m}$ , positioned above a vertical cylinder 10 with a diameter of 50  $\mu\text{m}$ . Powder is poured through the sieve, collected on top of horizontal face 11 of the cylinder 10 and then the maximum height of the cone of powder measured. According to the parameters of this test, the specification page 7 line 35 et seq. According to this test, the mobility of the powder increases as the height of the cone 12 decreases.

## **ISSUES**

Is Claim 13 unpatentable under 35 U.S.C 112 because of its definition of e.g.,  $D_{50}$ .

Are Claims 1-2, 4-10, 12 and 13 unpatentable for obviousness type double patenting over Claims 1-14 of U.S. Patent No. 6,171,567 in view of the English translation of DE 41 00 645A1?

Are Claims 1-2, 4-10, 12 and 13 unpatentable under 35 U.S.C. 103(a) as being unpatentable over German Patent Doc., No. DE 41 00 645 A1 to Regler et al in view of WO 95/19835 to Fagiolini?

## **GROUPING OF CLAIMS**

Claim 13 is separately patentable from the remaining claims since it is clear from the various grounds of rejection that the art does not pertain to the recitations in Claim 13.

## ARGUMENT

### I. Section 112 rejection of CLAIM 13.

In the Final Action, the reason advanced for the rejection of Claim 13 follows:

“Claim 13 is rejected under 35 U.S.C. 112, first paragraph, as containing subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventor(s), at the time the application was filed , had possession of the claimed invention. Neither specification on pg. 3 ln. 24 to pg. 4 ln. 4 or in claim 13 explain how  $D_{90}$  can represent the diameter at which 90% of the particles have a diameter less than  $D_{90}$ . It seems that one would have to know what the  $D_{90}$  is on the right side of “represents” in order to determine the  $D_{90}$  on the left side of “represents”. A similar rejection is made for the corresponding  $D_{50}$  and  $D_{10}$  expressions. [Paper No. 17, pages 2-3]

Contrary to the PTO reasons, the expressions are supported, i.e. appear at page 3 lines 19 through page 4 line 4 of the specification; that is, *in haec verbis* support was provided in the U.S. application specification for the expression(s). Specifically, at page 3 line 19 et seq. the specification recites,

In this embodiment of the invention, the mean diameter ( $D_m$ ) and the particle size slope ( $\sigma$ ) are defined by the following relationships:

$$D_m = [\sum n_i \times D_i] / \sum n_i \text{ [sic]}, \quad \sigma = [D_{90} - D_{10}] / D_{50}$$

In which  $n_i$  denotes the frequency (by weight) of the particles of diameter  $D_i$ , and  $D_{90}$  represents the diameter at which 90%...of the particles of the

reactive composition... have a diameter of less than

D90..."[specification,pg.4]

Moreover, on page 4 of the specification the appellants indicate the conventional means for making the measurement for particle classification.

Thus, on the strictest reading of Section 112, the PTO position appears inapposite, as both the written description requirement and enablement have been provided by the specification. However, to show on the record that the particle size classification was well within the skill of the art applicants presented various references which speak to particle size classification: Copies of the three references are attached hereto. These references date from 1986, 1975 and 1997.

Those previously submitted references include:

a page of Terence Allen's book entitled "Particle size Measurement", Second Edition, published by Chapman and Hall Ltd.(1975)[Enclosure 1] and

a copy of a page from Ullmann's, Encyclopedia of Industrial Chemistry, Fifth Edition, Vol. A7, p. 160-161, 1986, editor Wolfgang GERHARTZ, publisher:VCH.]

Those two references were attached to the two responses to the FINAL REJECTION. On October 1, 2002 appellants also filed

**Kirk Othmer Encyclopedia of Chemical Technology**, Fourth Edition, volume 22, Wiley Interscience Publication, p.276. [1997], including copyright page.

Better than an Inventor's declaration, this latter excerpt presents a very clear illustration of the "D90" etc concept: At the top of the page, a very clear cumulative graph is presented. To find, say, D90, you select 90 on the "Y" axis and read the corresponding D90 value on the "X" axis (idem for D50 etc.)



The totality of the references suggests the recitations relating to D90 , D50 and D10, would be well within the comprehension of a person of ordinary skill.

In conjunction with this position relating to the person of ordinary skill, the Board may wish to review applicants' Belgian representatives reference to D<sub>50</sub>, as set forth in Section IIB, below.

**Accordingly, reversal of the rejection under 35 U.S.C. 112 is respectfully solicited.**

**II. The claims are not obvious under 35 U.S.C. 103(a)**

**A. Claims 1, 2 and 4-12**

In Applicants' view, the references commend to the person of Section 103(a) inclusion of a reagent which is expressly eliminated from Claims 1 and 12, from which the remaining claims depend. [As noted in a previous response, PTO policy as expressed in the MPEP, Section 2111.03, the phrase "consisting essentially of" in e.g. appealed Claim 12 is construed to limit the scope of a claim to the specified materials and "those materials that do not materially affect the basic and novel characteristic(s)" of the claimed invention . In re Herz (citation omitted).] Elimination of an element suggested in the prior art with improved results in an area unsuggested by the prior art is the epitome of non-obviousness. Please see the cases, In re Anthony 64 USPQ 553 at 555-556 (CCPA 1945); In re Miller 94 USPQ 88 (CCPA 1952) ;In re Fleissner, 121 USPQ270 (CCPA 1959).

Applicants respectfully traverse the rejections of the claims under 35 U.S.C. 103(a) over German Patent No. DE 4100645 (Regler) which discloses a composition for the purification of gas, taken with WO 95/19835. The title of the Regler reference is "*Waste gas purificn. With nitrogen basic cpds. Removing acid cpds. – by adding ammonia and alkali and/or alkaline*

*earth cpds., for foundry, alkali chloride electrolysis, blast furnace, power station, refuse and glass industry.*” In general the Regler composition comprises:

- A basic alkaline and /or alkaline earth substance;
- A basic compound comprising nitrogen for absorption of NO<sub>x</sub>;
- An additive with large specific area, **including explicitly silica to absorb** certain impurities and improving the reactivity with the gas.

More specifically, the Abstract of the Regler reference recites

“Nitrogen bases (IA) are injected above the dw pt. Of H<sub>2</sub> in addn. to basic alkali and/or alkaline earth cpds. (IB), mixed with the gas stream and reacted and the solids are sepd. In dust separators. Zeolites are used as surfactant (II) together with (IB). Pref. (IA) is NH<sub>3</sub>, ammonium salts, e.g. NH<sub>4</sub>Cl, urea and/or prim., sec. and/or tert. Amines, (NH<sub>3</sub> gas) (B) is NaOH, KOH, NaHCO<sub>3</sub>, Na<sub>2</sub>CO<sub>3</sub>, KHCO<sub>3</sub>, K<sub>2</sub>CO<sub>3</sub>, quicklime, Ca(OH)<sub>2</sub>, limestone, MgO, Mg(OH)<sub>2</sub> and/or MgCO<sub>3</sub>, as solid, soln. or suspension. (IB) may be mixed with (II) content of the (IB)/(II) mixt. Is 0.1-95,(0.5-50) esp. 1-10%.”

Applicants respectfully traverse the rejection of Claims 1-10 and 12 under 35 U.S.C. 103(a) as unpatentable over German Patent Doc DE 4100645A1 to Regler [hereinafter ‘Regler’] in view of WO 95/19835.

With respect to the contents of WO 95/19835 [combined with Regler] applicants note that the PTO relies upon it for the English translation of the ABSTRACT. The abstract of this reference suggests that the reactive composition for purifying flue gases comprises sodium bicarbonate and less than 2 wt % of sodium monocarbonate, with a particle size

distribution defined by an average particle diameter of less than 0.05mm. Such disclosure does not make up for the deficiencies of Regler with respect to the appealed Claims.

**The differences between Regler and the claims on appeal include the following information:**

- Regler's invention lies in the addition of *nitrogen containing compounds, in order to reduce the emission of nitrogen oxides*. This is not the object of applicants' invention to basic alkali and/or alkaline earth compounds; and Regler's invention is not particularly relevant to the object of the invention, which includes solving an agglutination problem, of sodium bicarbonate. Thus, a man skilled in the art who wants to solve an agglutination problem *would not view the Regler's paper at all and the PTO has provided no evidence to the contrary!*
- In applicants' invention the bicarbonate is the main active constituent whereas the magnesium compound is a caking inhibitor *additive*. This is emphasized in originally filed claim 3 [now incorporated into claim 1]. To the contrary, in Regler, the alkaline earth compound can be the main or only (and even *preferred*, see the example) active basic constituent for the gas purification.
- Regler's compositions comprise alkali and/or alkaline earth. The selection of sodium bicarbonate *and* a magnesium compound among 6 alkali and 4 alkaline earth (burned lime, calcium hydroxide, calcium carbonate and magnesium compound) amounts to the selection of two elements among a list of 10 elements, that is one among 90 (10 times 9). **Moreover, Regler recommends adding a surface active substance, including one which is silica.** The probability **avoiding** silica is 4/5, since silica is in a list of 5 elements. In summary, the reconstitution of the constituents of the invention through

multiple selections in the 3 Regler's lists, amounts to a selection of one element in a hundred!

- In order to elicit applicants' invention from Regler, the skilled man would have had, after the selection of one-among a hundred of possibilities, to **modify the proportions** of the selected constituents of the composition. Indeed, in Regler, the magnesium compound is possibly the single candidate of choice as the basic active substance for gas purification. In his only example, Regler discloses a composition consisting of calcium hydroxide only (which is in the same list as the magnesium compounds). By comparison, according to the invention, the magnesium compound is an additive, in proportions of at most 10% in weight!
- The comparison of the applicants' specification examples 7 (in accordance with the invention) and 8 (not in accordance with the invention) reveals the particularly interesting advantage of avoiding, according to the invention, the presence of silica in the reactive composition.
- In conclusion, the Examiner's reasoning is hindsight reconstruction of the Regler invention. The complexity of the selections and modifications needed to reconstitute the claimed subject matter proves that the invention is *nonobvious* over Regler.

The PTO selection from the choices provided by Regler requires applicants' own claimed subject matter as the record here does not establish that Regler's description provides express motivation to exclude silica or an expectation of success. To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second,

there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art, not in applicant's disclosure. *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438 (Fed. Cir. 1991).

Applicants' composition is non obvious for the following reasons:

1. Applicants' invention is intended to solve an agglutination problem (the composition is a caking inhibitor-see claim 1-). **Regler does not mention nor suggest an agglutination problem.** As a consequence, after reading the DE 4100645 document, a person skilled in the art would not make, among Regler's numerous compositions, the very particular selection which solves a problem not even mentioned in the document;
2. In order to reconstitute Applicants' invention from DE 4100645 document, a person skilled in the art must operate successive selections
3. Regler's composition can contain silica (silica is explicitly mentioned among the possible compositions), whereas the Applicants have discovered the negative effect of silica for the agglutination problem. Specifically, Applicants' claims recite that the composition is substantially devoid of silica.

## II B. CLAIM 13 IS INDEPENDENTLY PATENTABLE UNDER 35 USC 103(a)

In Paper No. 29, the Examiner has asserted against Claim 13 a ground of rejection under 35 U.S.C. 103 which had been lodged against claims 1,2, 4-12 [over German Patent No. DE 4100645 (Regler) which discloses a composition for the purification of gas, taken with WO 95/19835], but not Claim 13; please see page 6 of Paper No. 17. Applicants traverse and

incorporated by reference the remarks concerning the description of the references above, for the purposes of brevity.

Claim 13 qualifies claim 1 by relating the term “slope” to distributions of particle diameter(s).

Obviousness cannot be predicated on the unknown, as the patent Statute requires the application of “prior art” evidence to support a rejection under 35 U.S.C. 103. Claim 13 is independently patentable of the remaining claims because the Final Rejection relies upon no art which describes the Claim 13 relationship of distribution of particle size diameters, which in turn define the term “slope” in Claim 1. The only comment concerning the patentability of Claim 13 under 35 USC is in the paragraph bridging pages 9 and 10 of Paper No. 29. No differentiation of the terms in Claim 1 vis-à-vis the terms of Claim 13 are noted. No reference is relied upon to show that the qualification of slope is a requirement of a single prior art composition.

The person of ordinary skill would view applicants’ claims as suggested by applicants’ Belgian representatives, by the following comparison example, in conjunction with the ‘median’ concept. The median salary of a population is the salary such that half of the people of the population earn more and half of the people earn less than that salary. This is common knowledge. Median salary could be denoted  $S_{50}$ . A median diameter is denoted  $D_{50}$  and embraces exactly the same concept since it is the diameter at which half of the particles are smaller and half of the particles are bigger.

In applicants’ view, the applied art does not describe Claim 13 recitations and does not suggest that requirement of compositions of Claim 13, to a person of ordinary skill. In applicants’ view, if a suggestion of granulometry is described in the WO 95/19835 reference as

alleged at page 10 of Paper NO. 29, this alleged suggestion does not 'suggest' claim 13 recitations.

Accordingly, reversal of the rejection of Claim 13 under 35 U.S.C. 103 is respectfully solicited.

**Reversal of the rejection under 35 U.S.C. 103(a) is respectfully solicited.**

**III. The claims are patentable over the "Obviousness-type Double Patenting Rejection.**

Applicants respectfully traverse the rejection of claims 1-10, 12 and 13 under the judicially created doctrine of obviousness-type double patenting over claims 1-14 of U.S. patent No. 6171567B1. The question presented by this rejection is: Does citation of the claims of a reference --under 'obviousness type double patenting-- logically pertain to **appealed claims** which **exclude compositions**, the **use of which**, in accordance with the reference claims, would **infringe the reference claims**?

Applicants' respectfully traverse the obviousness-type double patenting rejection. In applicants' view, the precepts of In re Vogel apply in the instant situation [In re Vogel, 422F2d 438, 164 USPT 619 (CCPA 1970)]; the issue of double patenting present an analysis akin to the determination of infringement, a question of fact. Substantively, applicants note that the discussion above with respect to the non-obviousness of the invention over Regler, also moots the double patenting rejection. In applicants' view combining two references which relate to inventions different from each other and different from the claims at issue does not provide a **prima facie** grounds for double patenting. Evidence of this is that the very composition which applicants' expressly seek to exclude from the claims at issue could be used in a way to infringe the claims of the cited patent [6171567B1].

Legally, applicants' note the Studiengesellschaft Kohle mbH v. Northern Petrochemical Co. [ 228 USPQ 837 Fed.Cir. 1986 ] case precedent. In this case, the court held that claims to a product [as claims 1 and 12 in the instant appeal] are not directed to the same invention as claims directed to a process of use, viz., "[B]ecause the two patents claim different statutory classes of subject matter, composition and process, they are not the same invention. Studiengesellschaft Kohle mbH v. Northern Petrochemical Co. [ 228 USPQ 837, at 840]"

In applicants' view combining two references which relate to inventions different from each other and different from the claims at issue does not provide a **prima facie** grounds for double patenting. Evidence of this is that the very composition which applicants' expressly seek to exclude from the claims at issue could be used in a way to infringe the claims of the cited patent [6171567B1].

#### CONCLUSION

**Reversal of the three grounds of rejection appears to be in order.**

Respectfully submitted,

Date:

*April 16 2003*

*Marina V. Schneller*

Marina V. Schneller

Registration No. 26,032

**Venable, Baetjer, Howard & Civiletti, LLP**

Post Office Box 34385

Washington, D.C. 20043-9998

Phone: (202) 962-4800

Fax: (202) 962-8300



## APPENDIX

### APPENDIX---CLAIMS

ok 1. Solid pulverulent reactive composition for the purification of a gas, comprising at least 90% by weight of sodium bicarbonate and a caking inhibitor for sodium bicarbonate and being devoid of silica, said inhibitor comprising lignite coke and/or a magnesium compound selected from the group consisting of magnesium oxide, magnesium hydroxide, mixtures of magnesium oxide and magnesium hydroxide and magnesium hydroxycarbonate, wherein said composition exhibiting a mean particle size of less than 50  $\mu\text{m}$  and a particle size slope of less than 5 and wherein said inhibitor is present in an amount by weight of greater than 0.5% of the weight of sodium bicarbonate.

ok 2. (Thrice Amended) The composition according to Claim 1, wherein said magnesium compound is magnesium hydroxycarbonate.

ok 4. The composition according to claim 3, wherein the inhibitor comprises a magnesium compound in an amount by weight at least equal to 2% of the weight of sodium bicarbonate.

ok 5. The composition according to Claim 3, wherein the inhibitor comprises lignite coke in an amount at least equal to 5% of the weight of sodium bicarbonate.

ok 6. A process for the purification of a gas, comprising introducing a reactive composition of Claim 1 into the gas and subjecting the gas to removal of dust.

ok 7. The process according to Claim 6, wherein said subjecting the gas to removal of dust comprises filtrating the gas through a filter cloth.

8. The process according to Claim 6, for the purification of a gas from at least one contaminant selected from the group consisting of hydrogen chloride, hydrogen fluoride, sulfur oxides, nitrogen oxides, dioxins and furans.

9. (Twice amended) The process according to Claim 7, wherein the reactive composition is that of Claim 2.

10. The process according to Claim 1 wherein said silica is one containing less than 0.48% of silica.

12. A non-caking solid pulverulent reactive composition for the purification of a gas containing HCl, HF, sulfur oxide, nitrogen oxide, dioxins, furans, and admixtures thereof, consisting essentially of

sodium bicarbonate and

a caking inhibitor for sodium bicarbonate,

said inhibitor is selected from the group consisting of lignite coke, a magnesium compound and admixtures thereof, wherein said magnesium compound is selected from the group consisting of magnesium oxide, magnesium hydroxide, mixtures of magnesium oxide and magnesium hydroxide, and magnesium hydroxycarbonate;  
wherein said composition is devoid of silica.

13. [Proposed] The [process] composition of Claim 1 wherein the particle size slope is defined by  $\sigma$ , wherein

$$\sigma = \frac{D_{90} - D_{10}}{D_{50}}$$

wherein  $D_{90}$  represents the diameter at which 90% of the particles of the reactive composition (expressed by weight) have a diameter of less than  $D_{90}$ ;

wherein  $D_{50}$  represents the diameter at which 50% of the particles of the reactive composition (expressed by weight) have a diameter of less than  $D_{50}$ ; and

wherein  $D_{10}$  represents the diameter at which 10% of the particles of the reactive composition (expressed by weight) have a diameter of less than  $D_{10}$ .

KIRK-OTHMER

# ENCYCLOPEDIA OF CHEMICAL TECHNOLOGY

FOURTH EDITION

VOLUME 22

SILICON COMPOUNDS  
TO  
SUCCINIC ACID AND SUCCINIC ANHYDRIDE



A Wiley-Interscience Publication  
**JOHN WILEY & SONS**

New York • Chichester • Weinheim • Brisbane • Singapore • Toronto

EDITOR  
Kroschwitz

EDITOR  
owe-Grant

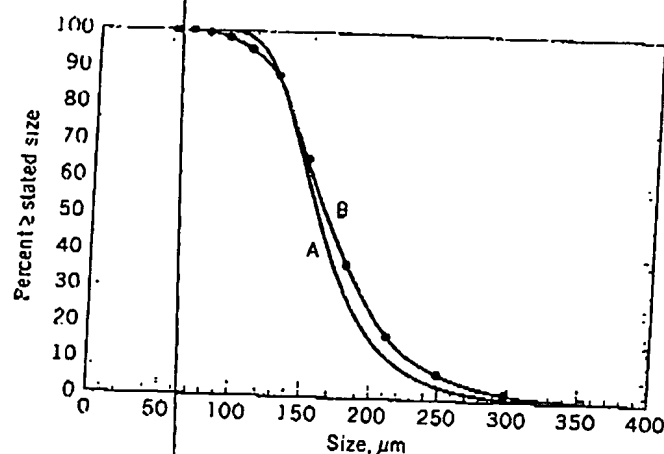


Fig. 15. Size data for a metal powder obtained by A, image analysis, and B, on a diffractometer.

Optical counters have been widely used to monitor cleanroom technology and particles in oil. Instruments manufactured by Royco Inc. (Menlo Park, California) are available for studying aerosols and particles in liquids. The HIAC counter (HIAC Instruments, Monte Claire, California) is a widely used stream counter for particles in fluid. One of the more recently developed optical counters is available from Particle Sizing Systems (Santa Barbara, California). The configuration of one of the widely used counters, the Climet counter, is shown in Figure 16. A general review of photozone counters is available (3).

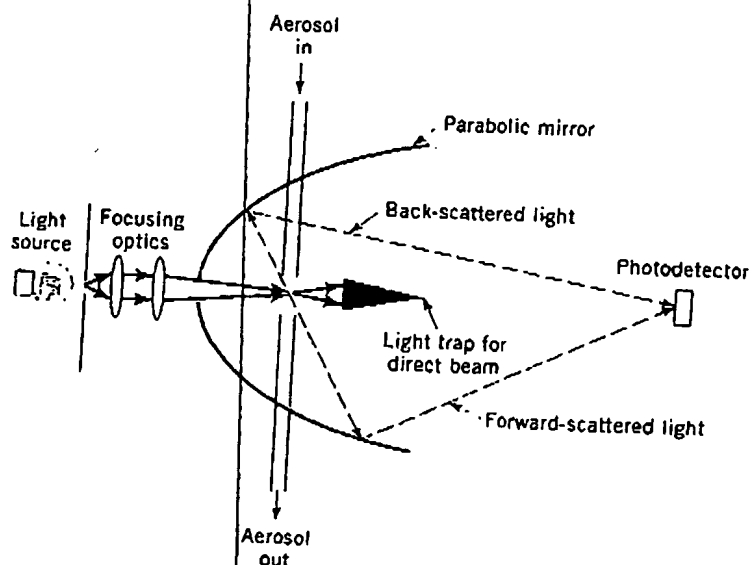


Fig. 16. Schematic representation of the internal structure of a Climet counter.

## BIBLIOGRAPHY

"Size Measurement of Particles," University of Minnesota, pp. 106-131, C.

1. R. A. Mugele, University of Minnesota, pp. 106-131, C.
2. Malvern Friction, Mass.,
3. B. H. Kaye, York, 1981.
4. Technical data, eachrome Co.
5. C. H. Murphy, ers, Weinhe
6. K. Sommer, 1986, p. 291.
7. British Standard, division of G, London, 196
8. T. Allen, Pa
9. B. H. Kaye, 194-197 (A)
10. B. H. Kaye, Mixer System, 5-7, 1996.
11. K. Leschonski
12. J. Hidaka ar
13. K. Schonert,
14. B. H. Kaye,
15. B. H. Kaye,
16. R. W. Bartle
17. C. Orr, D. K
18. A. Rudolph,
19. K. T. White, Publication
20. J. E. Englis
21. H. O. Suhm
22. C. W. Ward,
23. B. J. Wahl,
24. J. D. Zwicke
25. H. B. Carro
26. H. H. Heyw
27. H. H. Haus
28. T. Allen, Pa
29. G. Martin,
30. L. R. Feret
31. J. S. Glass,
32. J. C. Russ, North Caro
33. A. M. Glaue, the Netherl
34. N. Thaulow

This text is printed on acid-free paper.

Copyright © 1997 by John Wiley & Sons, Inc.

All rights reserved. Published simultaneously in Canada.

Reproduction or translation of any part of this work beyond that permitted by Sections 107 or 108 of the 1976 United States Copyright Act without the permission of the copyright owner is unlawful. Requests for permission or further information should be addressed to the Permissions Department, John Wiley & Sons, Inc., 605 Third Avenue, New York, NY 10158-0012.

#### Library of Congress Cataloging-in-Publication Data

Encyclopedia of chemical technology/executive editor, Jacqueline

I. Kroschwitz; editor, Mary Howe-Grant.—4th ed.  
p. cm.

At head of title: Kirk-Othmer.

"A Wiley-Interscience publication."

Contents: v. 22, Silicon compounds to succinic acid and succinic anhydride  
ISBN 0471-52691-6 (v. 22)

I. Chemistry, Technical—Encyclopedias. I. Kirk, Raymond E.

(Raymond Eller), 1890–1957. II. Othmer, Donald F. (Donald

Frederick), 1904–1995. III. Kroschwitz, Jacqueline I., 1942–

IV. Howe-Grant, Mary, 1948– V. Title: Kirk-Othmer encyclopedia  
of chemical technology.

TP9.E685 1992

91-16789

660.03—dc20

Printed in the United States of America.

10 9 8 7 6 5 4 3 2 1

Silicon Co  
Silk

Silver and  
Silver Cor

Simultane  
Transfe

Size Enla

Size Mea

Size Red

Soap

Sodium a

Sodium C

Soil Chem

Soil Stab

Solar Ene

Solders a

Metals

Sol-Get

Solvents

Sorbic Ac

Table 4.8 (a) Cumulative percentage undersize distribution

Particle size ( $\mu\text{m}$ )	Cumulative percentage undersize
$x_1$	$\phi = \sum_{n=1}^{\infty} u_n \phi$
5	1.4
9	9.4
11	18.0
14	32.0
17	49.5
20	64.0
23	76.0
28	88.0
33	94.0
41	98.0
50	99.4
60	99.9

$\phi$ , the frequency function =  $\sum u_n \phi$  for a number distribution  
 =  $\sum x_i^3 dN$  for a size distribution  
 =  $\sum x_i^3 dN$  for an area distribution  
 =  $\sum x_i^3 dN$  for a volume or weight distribution where  $u_n$   
 is the percentage of the total number of particles lying in the size range  $x_1$  to  $x_2$ .

Table 4.8 (b) Relative percentage frequency distribution: tabular calculation of mean size

Particle size range $x_1$ to $x_2$	Interval $\Delta x$	Average size $\bar{x}$	Percentage in range $\Delta \phi$	Percentage per micrometre $\phi/\Delta x$	$x \Delta \phi$
0 to 5	5	2.5	1.4	0.3	4
5 to 9	4	7.0	8.0	2.0	56
9 to 11	2	10.0	8.6	4.3	86
11 to 14	3	12.5	14.0	4.7	175
14 to 17	3	15.5	17.5	5.8	271
17 to 20	3	18.5	14.5	4.8	268
20 to 23	3	21.5	12.0	4.0	258
23 to 28	5	25.5	12.0	2.4	306
28 to 33	5	30.5	6.0	1.2	183
33 to 41	8	37.0	4.0	0.5	148
41 to 50	9	45.5	1.4	0.2	64
50 to 60	10	55.0	0.5	0.1	25
					$\sum x \Delta \phi = 1847$
					Mean size = $\frac{\sum x \Delta \phi}{\sum \Delta \phi} = 18.47$

PARTICLE SIZE, SHAPE AND DISTRIBUTION

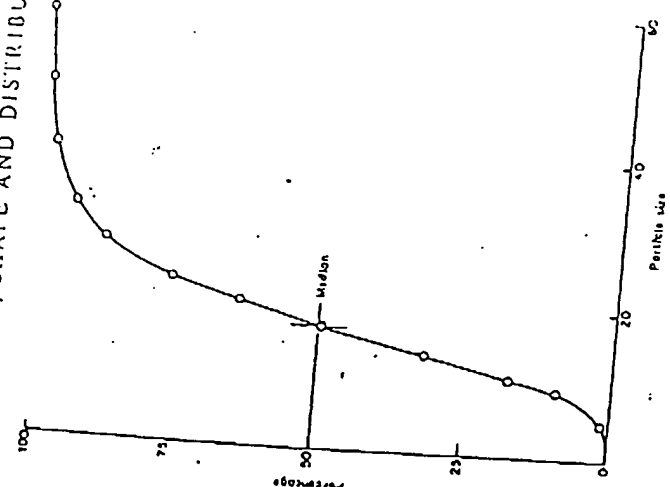


Fig. 4.3. The cumulative percentage frequency curve

passes through the centre of gravity of a sheet of uniform thickness and density cut to the shape of the distribution. Hence, for the mean, the moment of the sum of all the elementary areas of thickness  $\delta x$  about the ordinate equals the sum of all the moments:

$$\bar{x} \sum \frac{d\phi}{dx} \delta x = \sum x \frac{d\phi}{dx} \delta x$$

$$\bar{x} = \frac{\sum x d\phi}{\sum d\phi}$$

For a weight distribution  $d\phi = x^3 dN$  giving:

$$\bar{x} = \frac{\sum x^4 dN}{\sum x^3 dN}$$

The mode and the median may be determined graphically but the above summation has to be carried out for the determination of the mean. However, for slightly skewed distribution, the approximate relationship mean-mode = 3 (mean-median) holds. For a symmetrical distribution, they all coincide. In the illustration, the values are: mode = 15.0; median = 17.2; yielding mean = 18.2, as compared with the estimated value of 18.47 (table 4.8).

Enclosure 1

(4.13)

(4.14)



Figure 1. Geographic locations of the world's coal

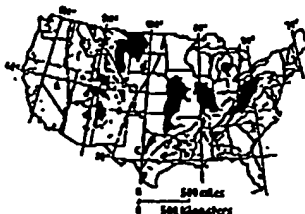


Figure 2. Coal provinces of the conterminous United States  
1) Eastern; 2) Gulf; 3) Interior; 4) Northern Great Plains; 5) Pacific Coast; 6) Rocky Mountains

younger, these coals tend to be of lower rank, usually subbituminous, than the Carboniferous coals. Since the Carboniferous coals have been deposited in scattered locations more or less continuously coal tends to be lignite or brown coal.

The distribution of coal seams throughout the world is also not uniform. As shown in Figure 1, most of the world's coal is located in only three countries, the United States, the Soviet Union, and China. Although the figures vary from source to source, each of these countries has about 25% of the total coal resources, while the rest of the world shares the remaining 25%. In the United States, bituminous coal seams are concentrated in the Appalachians and Illinois Basins. Most of the subbituminous coal occurs in the various smaller basins in the Rocky Mountain region, and the lignite seams are concentrated in the northern Great Plains and the Gulf Coast area.

#### 4. Classification

Coal is combustible and should be composed of more than 50 wt% carbonaceous material [11]. Commercially, coal is classified in a number of ways on the basis of (1) the original plant or mineral composition, sometimes called coal type, (2) the degree of maturity or metamorphism, called coal rank, (3) the amount of impurities such as ash or sulfur, called coal grade, and (4) the industrial properties such as caking or agglomeration.

One of the main classifications by composition used by the United States Bureau of Mines is based on the relative amounts of petrographic entities deposited in this section analysis, including anthracylane (translucent material roughly equivalent to vitrinite), transparent vitrinite (roughly equivalent to Epiptene), and opaque matter and fusinite (roughly equivalent to liptodite) [12], [13]. Under this system, coals are divided into two groups: **hard coals**, with > 5% anthracylane, and **subhard coals**, with < 5% anthracylane. The **hard coals** are subdivided into three types: **bright coal**, consisting mainly of anthracylane and transparent vitrinite with < 20% opaque matter; **semibright coal**, consisting mainly of transparent and opaque matter with 20–30% opaque matter; and **opaque coal**, consisting mainly of opaque matter with > 30% opaque matter. The **subhard coals** are divided into **semi-bright coal**, consisting of vitrinite with sparse, and **lignite coal**, consisting of lignite with sparse.

The various bands or layers in coal evident to the unaided eye have also been classified into four types [14]. **Fusinite** layers appear bright in vitrinite, **clastic** layers appear relatively less bright, **lignite** layers are dull gray and the **charcoal**, although these terms (all ending in -ite) are used as a guide to the eye, they do not have any compositional implications at the microscopic level. For example, vitrinite layers contain mainly vitrinite macerals, fusinite layers contain mainly fusinite macerals, and clastic and lignite are mixed with all three maceral types.

The most important classification for commercial purposes in the United States is the ASTM classification by rank. It is the basis of which most of the coal in the United States is bought and sold. This classification, ASTM Standard D 388 shown in Table 3, divides coal

Table 3. Classification of coal by rank\*

Class	Group	Fixed carbon basis, % (dry, mineral-matter-free basis)	Volatile matter basis, % (dry, mineral-matter-free basis)	Calorific value Btu/lb (metric kilocalories/kg)	Agglomerating character
Anthracite	metastable	≥ 86	≤ 12	≥ 13,000	non-agglomerating
	asthenic	≥ 82	≤ 14	≥ 12,500	
	subasthenic	≥ 78	≤ 16	≥ 12,000	
Semi-anthracite	low-volatile bituminous coal	≥ 78	≤ 14	≥ 12,000	agglomerating
	medium-volatile bituminous coal	≥ 74	≤ 16	≥ 11,500	
	high-volatile A bituminous coal	≥ 70	≤ 18	≥ 11,000	
	high-volatile B bituminous coal	≥ 66	≤ 20	≥ 10,500	
Subbituminous	subbituminous A coal	≥ 62	≤ 22	≥ 10,000	agglomerating
	subbituminous B coal	≥ 58	≤ 24	≥ 9,500	
	subbituminous C coal	≥ 54	≤ 26	≥ 9,000	
Lignite	lignite A	≥ 40	≤ 30	≥ 8,000	non-agglomerating
	lignite B	≥ 30	≤ 40	≥ 7,000	

\* This classification does not include a few coals, principally subbituminous and lignite, which have unusual physical and chemical properties and which come within the limits of fixed carbon or volatile matter of the high-volatile bituminous and subbituminous ranks. All of these coals either contain < 1% dry, mineral-matter-free fixed carbon or have > 13,500 Btu/lb metric kilocalories/kg.

\* Lignite refers to coal containing its natural inherent moisture but not including visible water on the surface of the coal.

\* If agglomerating, classify in low-volatile group of the bituminous class.

\* Coals having 60% or more fixed carbon on the dry, mineral-matter-free basis shall be classified according to fixed carbon, regardless of volatile matter.

\* It is recognized that there may be some agglomerating varieties in these groups of the lignite class, and there are notable exceptions in high-volatile C bituminous group.

into 4 classes, anthracite, bituminous, subbituminous, and lignite, which are further subdivided into 13 groups on the basis of fixed carbon and volatile matter content, calorific value, and agglomerating character. The fixed carbon and volatile matter values are on a dry, mineral-matter-free basis and the calorific values are on a moist, mineral-matter-free basis. In this system, coals with ≥ 60% fixed carbon are classified by fixed carbon content and those with < 60% fixed carbon are classified by calorific value. Thus, anthracite and subbituminous coals and the low-rank bituminous coals are classified by their calorific value. It is also important to note that not all coals can be fixed into this system. This is especially true of coals with a high lignite content and content, such as coking and boghead types.

The other important classification system is the international system of the ISO. In this system, coals are divided into two types: **hard coals**, with greater than 13.8% C/H<sub>2</sub> (10,260 Btu/lb) and **soft coals**, with less than 13.8% C/H<sub>2</sub> (10,260 Btu/lb). In the hard coal classification shown in Table 4, the coals are divided

into classes, groups, and subgroups. The classes are similar to ASTM groups and based on dry, ash-free volatile matter (p. 260) and moist, ash-free calorific value. The classes are numbered as 0 to 9. The classes are divided into four groups, numbered 0 to 3 on the basis of the swelling properties (free-swelling index (p. 248), also called oreable swelling number, and Ragi index). These groups are further broken down into six subgroups numbered 0–5 on the basis of their Andrusch-Aren distribution number and Gray-King color type. The system is set up in such a way that all coals are classified with a three-digit number, in which the first digit is the class, the second digit is the group, and the third digit is the subgroup.

The lignites and brown coals are only divided into classes and groups. The classes, numbered from 1 to 4, are based on ash-free moisture; the groups, based on dry, ash-free yield, are numbered from 0 to 4. This classification is shown in Table 5 (p. 163).

Although the ASTM and International Systems are different, there is a reasonable correlation